

In memory of my father

DUALITY AND EFFECTIVE CONDUCTIVITY OF TWO-DIMENSIONAL TWO-PHASE SYSTEMS

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The possible functional forms of the effective conductivity σ_{eff} of the randomly inhomogeneous two-phase system at arbitrary values of concentrations are discussed. A new functional equation, generalizing the duality relation, is deduced for systems with a finite maximal characteristical scale of the inhomogeneities and its solution is found. A hierarchical method of the construction of the model random inhomogeneous medium is proposed and one such simple model is constructed. Its effective conductivity at arbitrary phase concentrations is found in mean field like approximation. The derived formulas (1) satisfy all necessary inequalities and symmetries, including a dual symmetry; (2) reproduce the known formulas for σ_{eff} in weakly inhomogeneous case. It means that in general σ_{eff} of the two-phase randomly inhomogeneous system may be a nonuniversal function and can depend on some details of the structure of the randomly inhomogeneous regions. The percolation limit of the randomly inhomogeneous two-phase systems is briefly discussed.

The electrical transport properties of the disordered systems have an important practical interest. For this reason they are intensively studied theoretically as well as experimentally. In this region there is one classical problem about the effective conductivity σ_{eff} of inhomogeneous (randomly or regularly) heterophase system which is a mixture of $N(N \geq 2)$ different phases with different conductivities $\sigma_i, i = 1, 2, \dots, N$. We confine ourselves here by the simplest case of the two-dimensional heterophase systems with $N = 2$. Despite of its relative simplicity only some few general exact results have been obtained so far. Firstly, there is a general expression for σ_{eff} in case of weakly inhomogeneous isotropic medium, when the conductivity fluctuations $\delta\sigma$ are smaller than an average conductivity $\langle\sigma\rangle$ ($\delta\sigma \ll \langle\sigma\rangle$) similar to the analogous expression for the permitivity of the corresponding dielectric mixture [1]

$$\sigma_{eff} = \langle\sigma\rangle \left(1 - \frac{\langle\sigma^2\rangle - \langle\sigma\rangle^2}{D\langle\sigma\rangle^2} \right), \quad (1)$$

where D is a dimension of the system. In our simplest case of two-dimensional two-phase system $\langle\sigma\rangle = x\sigma_1 + (1-x)\sigma_2$, $\langle\sigma^2\rangle - \langle\sigma\rangle^2 = x(1-x)(\Delta\sigma)^2$, where

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x is a concentration of the first phase, $\Delta\sigma = \sigma_1 - \sigma_2$, and the first formula takes a form

$$\begin{aligned}\sigma_{eff} &= \sigma_1 \left(1 - (1-x) \frac{\Delta\sigma}{\sigma_1} - \frac{x(1-x)(\Delta\sigma)^2}{2\sigma_1^2} \right) = \\ &\sigma_+ \left(1 + (x-1/2) \frac{\Delta\sigma}{\sigma_+} - \frac{x(1-x)(\Delta\sigma)^2}{2\sigma_+^2} \right),\end{aligned}\quad (1')$$

where $\sigma_+ = (\sigma_1 + \sigma_2)/2$.

Another general formula has been obtained for the case of a small concentration of one phase (for example, with a conductivity σ_2) [1]

$$\sigma_{eff} = \sigma_1 \left(1 - (1-x) \frac{2(\sigma_1 - \sigma_2)}{\sigma_1 + \sigma_2} \right), \quad (2)$$

where $1-x \ll 1$ is a small concentration of the second phase and a round form of the inclusions of this phase is suggested.

The further progress in the solution of this problem is connected with papers [2, 3]. It was shown there that there is a dual transformation transforming one phase into another. This transformation allows to find an exact formula for σ_{eff} in case of systems with equal concentrations of the phases $x = x_c = 1/2$ [3]

$$\sigma_{eff} = \sqrt{\sigma_1 \sigma_2}. \quad (3)$$

This remarkable formula is very simple and universal since it does not depend on the type of the inhomogeneous structure of the two-phase system. For systems with unequal phase concentrations the dual transformation gives a relation between effective conductivities at adjoint concentrations x and $1-x$ or in terms of a new variable $\epsilon = x - x_c$ ($-1/2 \leq \epsilon \leq 1/2$) at ϵ and $-\epsilon$

$$\sigma_{eff}(x, \sigma_1, \sigma_2) \sigma_{eff}(1-x, \sigma_1, \sigma_2) = \sigma_1 \sigma_2 = \sigma_{eff}(\epsilon, \sigma_1, \sigma_2) \sigma_{eff}(-\epsilon, \sigma_1, \sigma_2). \quad (4)$$

Eq.(4) means that a product of the effective conductivities at adjoint concentrations is an invariant. Due to this relation one can consider σ_{eff} only in the regions $x \geq x_c$ ($\epsilon \geq 0$) or $x \leq x_c$ ($\epsilon \leq 0$).

In the following papers this dual transformation was generalized on systems in magnetic field [5, 6, 7], polycrystals [3] and heterophase (with $N > 2$) systems [8, 9]. The formula (4) was also checked and improved for some regularly inhomogeneous systems (of the chess-board type) [10, 11].

However the main interest in this problem has a formula for the effective conductivity at arbitrary phase concentrations. Another question appears naturally in this case: is this formula also universal or it can depend on the structure of the two-phase system? We will give here a general analysis of the possible functional forms of the effective conductivity in the case of arbitrary phase concentrations and will show that, due to the dual symmetry, in some cases the corresponding effective conductivities can be found in the explicit form. Then we will introduce the finite maximal scale averaging approximation (FMSA-approximation),

deduce in the framework of this approximation a new equation, connecting the effective conductivities at different concentrations, and find its solution and its physical meaning. We will introduce also a new model of the random two-phase system which gives in the same approximation an effective conductivity, coinciding with one obtained by series expansion. All these results demonstrate that, in general, a formula for the effective conductivity may be nonuniversal even in the two-phase case. At the end of the paper we will briefly discuss some peculiarities of the percolation limit.

Let us start our investigation of the isotropic classical random two-phase system in the case of arbitrary concentrations with a general analysis. The basic equation is the Ohm law, connecting the local current $\mathbf{j}(\mathbf{r})$ and local electric field $\mathbf{E}(\mathbf{r})$,

$$\mathbf{j}(\mathbf{r}) = \sigma(\mathbf{r})\mathbf{E}(\mathbf{r}), \quad (5)$$

where $\sigma(\mathbf{r})$ is a local conductivity. It must be supplemented with the corresponding boundary conditions on the boundaries of two phases [1]

$$j_n^1 = j_n^2, \quad E_t^1 = E_t^2, \quad (6)$$

where n, t denote the normal and tangent components and 1, 2 correspond to different phases. The electric field $\mathbf{E}(\mathbf{r})$ is a curl-free field

$$\nabla \times \mathbf{E}(\mathbf{r}) = 0, \quad (7)$$

and the current field is a divergenceless field

$$\nabla \cdot \mathbf{j}(\mathbf{r}) = 0. \quad (8)$$

The effective conductivity σ_{eff} of the isotropic system can be defined as a proportionality coefficient between averaged values of \mathbf{j} and \mathbf{E} over the area of the system

$$\bar{\mathbf{j}} = \sigma_{eff}\bar{\mathbf{E}}, \\ \bar{\mathbf{j}} \equiv \int \mathbf{j}(\mathbf{r})d\mathbf{r}/S, \quad \bar{\mathbf{E}} \equiv \int \mathbf{E}(\mathbf{r})d\mathbf{r}/S \quad (9)$$

where S is an area of the system. Due to the linearity of the defining equations an effective conductivity of the random systems must be a homogeneous function of degree one of $\sigma_i, i = 1, \dots, N$. In our case $N = 2$ and it is convenient to use instead of $\sigma_i, i = 1, 2$ another combinations of σ_i : $\sigma_{\pm} = (\sigma_1 \pm \sigma_2)/2$. Then, introducing a new variable $z = \sigma_-/\sigma_+$, ($-1 \leq z \leq 1$), an effective conductivity can be represented in the following, symmetrical relatively to both phases, form

$$\sigma_{eff}(\epsilon, \sigma_+, \sigma_-) = \sigma_+ f(\epsilon, \sigma_-/\sigma_+) = \sigma_+ f(\epsilon, z), \quad (10)$$

where $\sigma_{eff}(\epsilon, \sigma_+, \sigma_-)$ and $f(\epsilon, z)$ must have the next boundary values

$$\sigma_{eff}(1/2, \sigma_+, \sigma_-) = \sigma_1, \quad \sigma_{eff}(-1/2, \sigma_+, \sigma_-) = \sigma_2, \\ f(1/2, z) = 1 + z, \quad f(-1/2, z) = 1 - z, \quad f(\epsilon, 0) = 1. \quad (10')$$

The duality relation, being a consequence of a duality of gradient and tangent fields in two dimensions [3], takes in these variables the form

$$f(\epsilon, z)f(-\epsilon, z) = 1 - z^2, \quad (11)$$

from which it follows that at critical concentration $\epsilon = 0$

$$f(0, z) = \sqrt{1 - z^2}. \quad (3')$$

Strictly speaking, this form of a duality relation is also a consequence of another exact relation for the effective conductivity, taking place at arbitrary concentrations for systems with the similar random structures of both phases of the system,

$$\sigma_{eff}(\epsilon, \sigma_1, \sigma_2) = \sigma_{eff}(-\epsilon, \sigma_2, \sigma_1). \quad (12)$$

It means that the effective conductivity of the random two-phase system must be invariant under substitution of these phases ($\sigma_1 \longleftrightarrow \sigma_2$) with the corresponding change of their concentrations $x \longleftrightarrow 1 - x$ (or $\epsilon \rightarrow -\epsilon$). In the new variables it means that

$$f(\epsilon, z) = f(-\epsilon, -z), \quad f(-\epsilon, z) = f(\epsilon, -z). \quad (13)$$

For this reason a duality relation can be written also in the form

$$f(\epsilon, z)f(\epsilon, -z) = 1 - z^2.$$

It follows from (13) that the even (f_s) and odd (f_a) parts of $f(\epsilon, z)$ relatively to ϵ coincide with the even (f^s) and odd (f^a) parts of $f(\epsilon, z)$ relatively to z

$$\begin{aligned} f^a(\epsilon, z) &\equiv \frac{1}{2}(f(\epsilon, z) - f(\epsilon, -z)) = f_a(\epsilon, z) \equiv \frac{1}{2}(f(\epsilon, z) - f(-\epsilon, z)), \\ f^s(\epsilon, z) &\equiv \frac{1}{2}(f(\epsilon, z) + f(\epsilon, -z)) = f_s(\epsilon, z) \equiv \frac{1}{2}(f(\epsilon, z) + f(-\epsilon, z)). \end{aligned} \quad (14)$$

The simplest way to satisfy (13) corresponds to the following functional form of $f(\epsilon, z)$

$$f(\epsilon, z) = f(\epsilon z, \epsilon^2, z^2) \quad (15)$$

One can conclude from (15) that in this case an expansion of $f(\epsilon, z)$ near the point $\epsilon = z = 0$ does not contain terms linear in ϵ and z separately. Moreover, one can reduce to the form (15) any function satisfying (13), since every combination of functions of two variables, odd relatively to the inversion of their arguments, can be made even by multiplying or dividing them by ϵz

$$\begin{aligned} F(-\epsilon, z) &= -F(\epsilon, z) = F(\epsilon, -z), \quad F(\epsilon, z) = \epsilon z \Phi(\epsilon^2, z^2), \\ \Phi(\epsilon^2, z^2) &= (\epsilon z)^{-1} F(\epsilon, z). \end{aligned}$$

Analogously, the odd part f_a can be represented in the form

$$f_a(\epsilon, z) = 2\epsilon z \Phi(\epsilon, z) \quad (16)$$

where Φ is an even function of ϵ and z (the coefficient 2 in front of ϵz is chosen for further convenience).

The duality relation (11) in general case is not enough for the complete determination of f , it only connects f_a and f_s

$$f_s^2 - f_a^2 = 1 - z^2. \quad (17)$$

It means that f_a and f_s considered at fixed z as the functions of ϵ satisfy to hyperbolic relation with a constant depending on z . The relation (17) allows to express $f(\epsilon, z)$ through its even or odd parts

$$f(\epsilon, z) = f_a + \sqrt{f_a^2 + 1 - z^2} = f_s \pm \sqrt{f_s^2 - 1 + z^2}. \quad (18)$$

For this reason it is enough to know only one of these two parts. Usually one prefers to choose an antisymmetric part as more simple. It follows from (1') that in the weakly inhomogeneous case the odd part has the simplest form (16) with $\Phi = 1$

$$f_a(\epsilon, z) = 2\epsilon z. \quad (19)$$

As is well known, the substitution of (19) into (18) gives an effective conductivity in the effective medium (EM) approximation [4]

$$\sigma_{eff}(\epsilon, z) = \sigma_+ \left[2\epsilon z + \sqrt{(2\epsilon z)^2 + 1 - z^2} \right], \quad (20)$$

This expression, being continued on arbitrary concentrations $\epsilon = x - 1/2$ and inhomogeneities z , reproduces in the corresponding limits both formulas (1') and (2).

Though at first sight a duality relation is not enough for finding f , usually systems with such symmetry have some additional hidden properties, permitting to obtain more information about function under question. Moreover, in some cases they can help to solve problem exactly (see, for example, [12]). Having this in mind, we will try to investigate the duality relation in more details. Since a homogeneous limit $z \rightarrow 0$ is a regular point of f it will be very useful to consider a series expansion of f in z

$$f(\epsilon, z) = \sum_0^\infty f_k(\epsilon) z^k / k!, \quad (21)$$

where due to boundary conditions (10')

$$f_0 = 1, \quad f_1(\epsilon) = 2\epsilon. \quad (22)$$

For every fixed z ($0 \leq z \leq 1$) a function f must be a monotonous function of ϵ . Substituting the expansion (21) into (11) one obtains the following results:

(1) in the second order in z it reproduces the universal formula (1'), thus the latter can be considered as a consequence of the duality relation;

(2) in general case there are the recurrent relations between f_{2k} and f_{2k-1} , corresponding to the connection (17);

(3) $f_{2k+1}(\epsilon)$ are odd polynomials of ϵ of degree $2k+1$ and $f_{2k}(\epsilon)$ are even polynomials of ϵ of degree $2k$ in agreement with (15).

Taking into account boundary conditions (10') and an exact value (3'), one can show that the coefficients f_k must have the next form

$$\begin{aligned} f_{2k+1}(\epsilon) &= \epsilon(1 - 4\epsilon^2)g_{2k-2}(\epsilon), \quad k \geq 1, \\ f_{2k}(\epsilon) &= (1 - 4\epsilon^2)h_{2k-2}(\epsilon), \quad k \geq 1, \end{aligned} \quad (23)$$

where g_{2k-2} and h_{2k-2} are some even polynomials of the corresponding degree and free terms of h_{2k-2} coincide with the coefficients in the expansion of (3')

$$\sqrt{1 - z^2} = 1 - z^2/2 - z^4/8 - z^6/16 - z^8/128 - z^{10}/256 + \dots \quad (24)$$

It follows from (23) that f_3 is completely determined up to overall factor number g_0 . Since f_4 is determined through lower f_k ($k = 1, 2, 3$)

$$f_4 = 4f_1f_3 - 3f_2^2 = (1 - 4\epsilon^2)[(8g_0 + 12)\epsilon^2 - 3], \quad (25)$$

it is also determined by the coefficient g_0 . The case $g_0 = 0 = g_{2k+1}$ ($k > 1$) corresponds to the EM approximation and in this case $f_{2k}(\epsilon) \sim (1 - 4\epsilon^2)^k$. Thus we see that in general case the arbitrariness of f is strongly reduced by boundary conditions and known exact value and that the third and fourth orders are determined only up to one constant. Then one can suppose that any additional information about function f can determine this constant or even the whole function. For this reason one can ask: what kind of functions can satisfy the duality relation (11)? In order to answer on this question it is convenient in the case $z \neq 1$ to pass from f to $\tilde{f} = f/\sqrt{1 - z^2}$. Then

$$\tilde{f}(\epsilon, z)\tilde{f}(-\epsilon, z) = 1 = \tilde{f}(\epsilon, z)\tilde{f}(\epsilon, -z). \quad (11')$$

The duality relation gives some constraints on the possible functional form of $\tilde{f}(\epsilon, z)$. For example, assuming a functional form (15), one can write out a simple function:

$$\tilde{f}(\epsilon, z) = \exp(\epsilon z \phi(\epsilon, z)), \quad (26)$$

where $\phi(\epsilon, z)$ is some even function of its arguments. It is easy to see that it automatically satisfies eq.(11'). Let us now consider two simple cases, when (a) $\phi(\epsilon, z)$ depends only on z , (b) it depends only on combination ϵz . Expanding the corresponding functions \tilde{f} in series one can check after some algebra that one can now determine all polynomial coefficients unambiguously! Another way to see this is to apply boundary conditions directly to the function (26). In the case (a) one obtains

$$\phi(z) = 1/z \ln \frac{1+z}{1-z}, \quad \tilde{f}(\epsilon, z) = \left(\frac{1+z}{1-z} \right)^\epsilon. \quad (27)$$

In case (b), when ϕ depends only on combination ϵz , one finds

$$\phi(\epsilon z) = \frac{1}{2\epsilon z} \ln \frac{1+2\epsilon z}{1-2\epsilon z}, \quad \tilde{f}(\epsilon, z) = \left(\frac{1+2\epsilon z}{1-2\epsilon z} \right)^{1/2}. \quad (28)$$

Series expansions of (27) and (28) coincide exactly with the corresponding expansions mentioned above. Unfortunately, analyzing an expansion of (26) under an assumption of regular behaviour of the coefficients $\phi_k(\epsilon)$ from a series expansion of $\phi(\epsilon, z)$

$$\phi(\epsilon, z) = \sum_0^{\infty} \phi_k(\epsilon) z^{2k} / k!$$

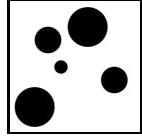
in ϵ

$$\phi_k(\epsilon) = \sum_0^{\infty} \phi_{kl}(\epsilon)^{2l} / l!,$$

one can show that now again the boundary conditions (10') (or (23)) cannot define all coefficients completely. For example, f_3 and f_4 contain one free parameter ϕ_{10} : $g_0 = 6(\phi_{10} - 1)$. It is very important to find any other solutions. Below we will present two simple models having the effective conductivity just of two forms found above.

Firstly, we will obtain an additional equation for effective conductivity using a new composite method, which we will call as a finite maximal scale averaging approximation. In framework of this method one divides an averaging procedure on two steps. This approximation can be considered as some modification of the mean field approximation and it is applicable to the inhomogeneous systems with a finite maximal scale of the inhomogeneities. Below in this section we will use temporally, for clarity, the concentration variable x instead of ϵ . Let us consider two-dimensional two-phase randomly inhomogeneous system. It is easy to see that, in general, its effective conductivity will depend on a scale l of a region over which an averaging is done. This takes place due to the possible existence of different characteristic scales in the inhomogeneous medium. In the most general case there will be a whole spectrum of these characteristic scales. This spectrum can be very different from discrete finite till continuous infinite and will define the inhomogeneity structure of the system. For this reason this spectrum can depend on concentration x . It is obvious that the effective conductivity of the system can depend on this spectrum as a whole as well as on which area an averaging is fulfilled over. Suppose, for the simplicity, that the randomly inhomogeneous structure of our system has the scale spectrum with a maximal scale $l_m(x)$, which is finite for all x in the region $1 \geq x > 1/2$ (or $1 - x$ in the region $0 \leq 1 - x < 1/2$). Let us assume that we know an exact formula for $\sigma_{eff}(x, z)$ of this system, which is applicable from scales $l > l_m$. It means that this formula for $\sigma_{eff}(x, z)$ takes place after the averaging over regions with a mean size $l \gtrsim l_m$ and does not change for all larger scales $l \gg l_m$. Now consider a square lattice with the squares of length $l_L \gg l_m$ and suppose that they have the effective conductivities corresponding to different values of the concentrations x_1 and x_2 with equal probabilities $p = 1/2$ (see Fig.1)

After the averaging over the scales $l \gtrsim l_L$ one must obtain on much larger scales $l \gg l_L$ the same effective conductivity, but corresponding to another concentration $x = (x_1 + x_2)/2$. This is possible due to the similar random structure of different squares and due to the conjectures that in this model:



(a)

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 1 | 2 | 1 | 1 | 2 | 2 | 1 | 1 | 2 |
| 2 | 1 | 2 | 2 | 2 | 2 | 1 | 1 | 2 |
| 1 | 2 | 1 | 2 | 1 | 2 | 1 | 1 | 2 |
| 2 | 1 | 2 | 2 | 1 | 2 | 2 | 1 | 2 |
| 1 | 2 | 1 | 1 | 1 | 2 | 1 | 1 | 1 |

(b)

Figure 1: (a) An elementary square of the model with round inclusions, (b) a lattice of the model, the numbers 1,2 denotes squares with the corresponding concentrations.

(1) there are only two maximal characteristic scales $l_m(x_i) \equiv l_i$ ($i = 1, 2$) and the lattice square size $l_L \gg l_i, l_m(x)$, (2) the averaging procedures over these scales do not correlate (or weakly correlate) between themselves. Thus for compatibility all concentrations must be out of small region around critical concentration x_c , where l_i or $l_m(x)$ can be very large. We will call further this set of the conjectures the finite maximal scale averaging approximation (FMSA approximation). It can be implemented for systems with compact inhomogeneous inclusions with finite l_m . From the other side the effective conductivity on scales $l \gg l_L$ must be determined by the universal Keller – Dykhne formula (3). Thus we obtain the next functional equation for the effective conductivity, connecting $\sigma_{eff}(x, z)$ at different concentrations,

$$\sigma_{eff}(x, z) = \sqrt{\sigma_{eff}(x_1, z)\sigma_{eff}(x_2, z)}, \quad x = (x_1 + x_2)/2, \quad (x_i, x \neq x_c). \quad (29)$$

It must be supplemented by the boundary conditions (10'). This equation can be considered as a generalization of the duality relation (4), the latter being a particular case of (29) at $x_1 + x_2 = 1$. It follows from (29) that, due to the exactness of the duality relation, it really works for all concentrations x , except maybe of small region near $x = x_c$ and $z = 1$ (this region corresponds to the singular region of the percolation problem, see also below a discussion of the percolation limit). One can easily check that σ_{eff} in low concentration limit (2) satisfies (29). Moreover, one can find an exact solution of this equation. It has an exponential form with a linear function of x (or ϵ)

$$\sigma_{eff}(x, z) = \sigma_1 \exp(ax + b), \quad (30)$$

where the constants a, b can be determined from the boundary conditions

$$a = -b, \quad \exp b = \sigma_2/\sigma_1. \quad (30')$$

Substituting these coefficients into (30) one obtains

$$\sigma_{eff}(x, z) = \sigma_1 (\sigma_2/\sigma_1)^{(1-x)} = \sigma_+ \sqrt{1-z^2} \left(\frac{1+z}{1-z} \right)^\epsilon. \quad (31)$$

The solution (31) satisfies all symmetry relations (4,11',13) and can be represented in the exponential form

$$\sigma_{eff}(x, z) = \sigma_+ \sqrt{1 - z^2} \exp \left(\epsilon \ln \frac{1+z}{1-z} \right), \quad (31')$$

which exactly coincides with the case (a) from (27) with $\phi(\epsilon, z) = \frac{1}{z} \ln \frac{1+z}{1-z}$. Its odd part has a form

$$\tilde{f}_a(\epsilon, z) = \sinh(\epsilon z \phi(\epsilon, z)), \quad (32)$$

and satisfies (17).

It is interesting to note that the form of the solution (31) means that in the considered approximation one has effectively an averaging of the $\log \sigma$ since it can be represented as

$$\log \sigma_{eff} = \langle \log \sigma \rangle = x \log \sigma_1 + (1-x) \log \sigma_2. \quad (33)$$

This coincides with a note made in [3] for the case of equal concentrations $x = 1/2$ and with analogous results obtained later for random system in the theory of the localization [13]. One can check that (31) reproduces in the weakly inhomogeneous limit the universal Landau – Lifshitz expression (1). In the low concentration limit of the second phase it gives

$$\sigma_{eff}(x, z) = \sigma_1 \left(1 + (1-x) \log \frac{1-z}{1+z} + \dots \right), \quad 1-x \ll 1, \quad (34)$$

what coincides with (2) in the weakly inhomogeneous case. Note that the expansion (34) contains the coefficients diverging in the limit $|z| \rightarrow 1$. Such behaviour of the coefficients denote the existence of a singularity in this limit (see below a discussion of this percolation limit).

Now we will show that an existence of the different functional forms for the effective conductivity denotes a nonuniversal character of this value for two-phase systems with different random structures. We will construct a model of two-dimensional isotropic randomly inhomogeneous two-phase system, using the composite method introduced above, and find a mean field like expression for its effective conductivity $\sigma_{eff}(x, z)$ in case of arbitrary phase concentrations x . The derived formula satisfies again to all necessary symmetries, including a dual one, and coincides with the example case (b) from (28), realizing the second variant, when $\tilde{f}(x, z)$ depends (in this approximation) only on one combination of variables ϵz and this dependence is described by the function analytical at small values of this variable.

Let us consider the following two-dimensional model. There is a simple square lattice with the squares consisting of a random layered mixture of two conducting phase with constant conductivities $\sigma_i, i = 1, 2$ and the corresponding concentrations x and $1-x$. A schematic picture of such square is given in Fig.2.

The layered structure of the squares means that the squares have some preferred direction, for example along the layers. Let us suppose that the directions



Figure 2: (a) An elementary square of the model with a vertical orientation, the dotted regions denote layers of the second phase; (b) a lattice of the model, the small lines on the squares denote their orientations.

of different squares are randomly oriented (parallelly or perpendicularly) relatively to the external electric field, which is directed along x axis. In order for system to be isotropic the probabilities of the parallel and perpendicular orientations of squares must be equal or (what is the same) the concentrations of the squares with different orientations must be equal $p_{||} = p_{\perp} = 1/2$.

Such lattice can model a random system consisting from mixed phase regions, which can be roughly represented on the small macroscopic scales as randomly distributed plots with the effective "parallel" and "serial" connections of the layered two-phase mixture (Fig.2). The lines on the squares denote their orientations.

This structure can appear, for example, on the intermediate scales when a random medium is formed as a result of the stirring of the two-phase mixture. The corresponding averaged parallel and perpendicular conductivities of squares $\sigma_{||}(x)$ and $\sigma_{\perp}(x)$ are defined by the following formulas

$$\begin{aligned}\sigma_{||}(x) &= x\sigma_1 + (1-x)\sigma_2 = \sigma_+(1+2\epsilon z), \\ \sigma_{\perp}(x) &= \left(\frac{x}{\sigma_1} + \frac{1-x}{\sigma_2}\right)^{-1} = \sigma_+ \frac{1-z^2}{1-2\epsilon z}.\end{aligned}\quad (35)$$

Thus we have obtained the hierarchical representation of random medium (in this case a two-level one). On the first level it consists from some regions (two different squares) of the random mixture of the two layered conducting phases with different conductivities σ_1 and σ_2 and arbitrary concentration. On the second level this medium is represented as a random parquet constructed from two such squares with different conductivities $\sigma_{||}$ and σ_{\perp} , depending nontrivially on concentration of the initial conducting phases, and randomly distributed with the same probabilities $p_i = 1/2$ (Fig.2). This representation allows us to divide the averaging process into two steps (firstly averaging over each square and then averaging over the lattice of squares) and implement on the second step the exact formula (3). This can be considered as some modification of

the FMSA approximation. As a result one obtains for the effective conductivity of the introduced random two-phase model the following formula, which is applicable for arbitrary concentration

$$\sigma_{eff}(\epsilon, z) = \sigma_+ \sqrt{1 - z^2} \tilde{f}(\epsilon, z), \quad \tilde{f}(\epsilon, z) = \left[\frac{1 + 2\epsilon z}{1 - 2\epsilon z} \right]^{1/2}, \quad (36)$$

This function has all necessary properties and satisfies equation (11') and symmetry (13). It coincides with the type (b) from (28) and has another possible functional form, automatically satisfying the duality relation (11')

$$\tilde{f}(\epsilon, z) = B(\epsilon, z)/B(-\epsilon, z) \quad (37)$$

with a function $B(\epsilon, z) = [1 + 2\epsilon z]^{\frac{1}{2}}$, which depends only on the combination ϵz .

It is interesting to compare this formula with the known general formulas. In order to do this one needs to find an asymptotic behaviour of the derived effective conductivity in different limiting cases. Let us consider firstly its behaviour for small phase concentrations.

(a) In case of small concentration of the first phase $x \ll 1$ one gets

$$\sigma_{eff}(x, z) \simeq \sigma_2 \left(1 + \frac{2xz}{1 - z^2} \right). \quad (38)$$

It follows from (38) that an addition of small part of the first higher conducting phase increases an effective conductivity of the system as it should be.

(b) In the opposite case of small concentration of the second phase $1 - x \ll 1$ one obtains

$$\sigma_{eff}(x, z) \simeq \sigma_1 \left(1 - \frac{2(1-x)z}{1 - z^2} \right), \quad (39)$$

i.e. an addition of the phase with smaller conductivity decreases σ_{eff} . It is worth to note that both these expressions for arbitrary values of the conductivities σ_1 and σ_2 differ from equation (2) and coincide with it only in the weakly inhomogeneous case $z \ll 1$. It must be not surprising because a form of the inclusions of the second phase in this model has completely different, layered, structure. In the low concentration expansion as well as directly in formula (36) one can see again that the divergencies appear in the limit $z \rightarrow 1$.

(c) In case of almost equal phase concentrations $x = 1/2 + \epsilon$, $\epsilon \ll 1$ one obtains

$$\tilde{f}(\epsilon, z) \simeq 1 + 2\epsilon z, \quad \sigma_{eff}(\epsilon, z) \simeq \sigma_+ \sqrt{1 - z^2} (1 + 2\epsilon z). \quad (40)$$

The Keller – Dykhne formula (3) is reproduced for equal concentrations.

The antisymmetric part of \tilde{f} has the following form

$$\tilde{f}_a(\epsilon, z) = \frac{\tilde{f}^2(\epsilon, z) - 1}{2\tilde{f}(\epsilon, z)} = \frac{2\epsilon z}{[1 - 4\epsilon^2 z^2]^{1/2}}. \quad (41)$$

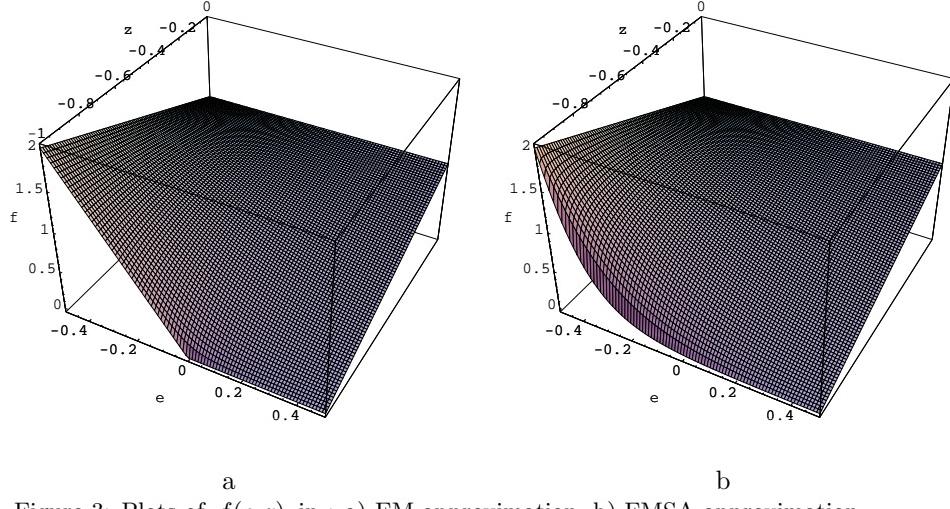


Figure 3: Plots of $f(\epsilon, z)$ in : a) EM approximation, b) FMSA approximation.

It follows from formula (41) that for $\epsilon \ll 1$ and (or) $z \ll 1$ the odd part f_a coincides in the first order with the corresponding expression from the effective medium theory. The corresponding function Φ is

$$\Phi(\epsilon, z) = \frac{\sqrt{1 - z^2}}{[1 - 4\epsilon^2 z^2]^{1/2}}. \quad (42)$$

In other words it is an analytic function of ϵz near $\epsilon z = 0$. Basing on this formula one can conjecture that an exact expression for $\tilde{f}_a(\epsilon, z)$ of this model will have a similar structure in general case with $\sigma_i \neq 0$ or $1 - z \neq 1$. One must note that at the same time the formula (36) does not satisfy the equation (29) except of the trivial case $x_1 = x_2$.

For the comparison of the different expressions of the effective conductivity we have constructed three-dimensional plots of $f(\epsilon, z)$ in the EM approximation, in the FMSA approximation and of the hierarchical model in FMSA-like approximation (fig.3,4).

It follows from these plots that all three formulas for σ_{eff} , despite of their different functional forms, differ from each other weakly for $z \lesssim 0,8$ due to very restrictive boundary conditions (10') and the exact Keller-Dykhne value. This range of z corresponds approximately to the ratio $\sigma_2/\sigma_1 \sim 10^{-1}$. For the smaller ratios a difference between these functions become distinguishable.

Now let us consider in the more details the derived formulas for $\sigma_{eff}(\epsilon, z)$ in case when $\sigma_2 \rightarrow 0(z \rightarrow 1)$. It is clear that for regularly inhomogeneous medium one can always construct such distribution of the conducting phase that $\sigma_{eff}(\epsilon, 1)$ will differ from zero for all $1/2 \geq \epsilon > -1/2$. But in the case of randomly inhomogeneous medium the limit $\sigma_2 \rightarrow 0$ is equivalent to the well known percolation problem [14,15]. In terms of z it corresponds to the limit

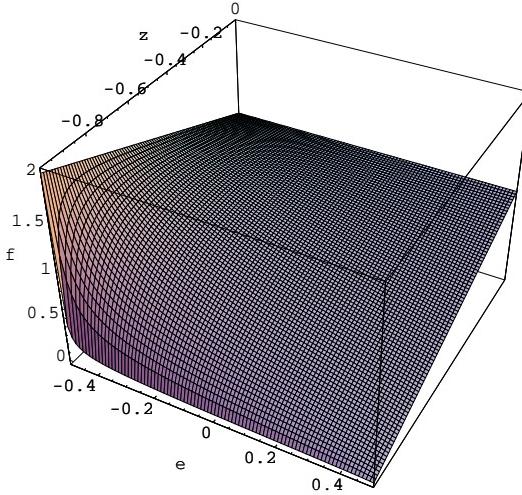


Figure 4: Plot of $f(\epsilon, z)$ of composite model in FMSA-like approximation

$z \rightarrow 1$ and is also similar to the superconducting limit $\sigma_1 \rightarrow \infty$. Strictly speaking, an implementation of the duality transformation (4) is not obvious in this case. However, if one supposes that the dual symmetry relation (4) fulfills in this limit too due to a continuity then it follows from (4) that

$$\sigma_{eff}(\epsilon)\sigma_{eff}(-\epsilon) = 0. \quad (43)$$

The relation (43) does not contradict to the known basic results of the percolation theory that $\sigma_{eff}(\epsilon) = 0$ for $\epsilon \leq 0$ and $\sigma_{eff}(\epsilon) \neq 0$ for $\epsilon > 0$. Moreover it follows from the general formula (17) that in this case $\sigma_s = |\sigma_a|$ and $\sigma_{eff}(\epsilon) = \sigma_a + |\sigma_a|$. It gives

$$\sigma_{eff}(\epsilon) = \begin{cases} 0, & \epsilon \leq 0, \\ 2\sigma_a, & \epsilon > 0. \end{cases} \quad (44)$$

This means that a behaviour of $\sigma_{eff}(\epsilon)$ in the percolation theory is completely determined by its odd part. From a general discussion of the functional structure of the conductivity of the two-dimensional randomly inhomogeneous systems and the known experimental and numerical results it is known that in the percolation limit the effective conductivity σ_{eff} must have a nonanalytical behaviour near the percolation edge $x_c = 1/2$ or at small $\epsilon > 0$

$$\sigma_{eff}(\epsilon) \sim \sigma_1(x - x_c)^t \sim \sigma_1 \epsilon^t, \quad (45)$$

where a critical exponent of the conductivity t is slightly above 1 and can be represented in the form $t = 1 + \delta$. Since the values of this exponent found by the numerical calculations are confined to be in the interval $(1.10 - 1.4)$ [14],

then δ have to be small and belongs to the interval $(0.1 - 0.4)$. It follows from general formula (44) that one must have

$$f_a(\epsilon, z) \xrightarrow[z \rightarrow 1, \epsilon \rightarrow 0_+]{} \epsilon^t. \quad (46)$$

It means that the function $\Phi(\epsilon, z)$ has at small ϵ some crossover on z under $z \rightarrow 1$ from a regular (analytical) behaviour to a singular one. At the moment an exact form of this crossover is unknown. For example, it can be of the form

$$\Phi(\epsilon, z) \sim (\Phi_0(z^2) + \Phi_1(z^2)\epsilon^2)^{\delta(z)/2}, \quad z \rightarrow 1, \quad (47)$$

where $\Phi_0(z^2) \rightarrow 0$, $\Phi_1(z^2) \rightarrow \Phi_1 \neq 0$ and $\delta(z) \rightarrow \delta \neq 0$ when $z \rightarrow 1$.

However, as it follows from the formulas obtained above, one gets always $\sigma_{eff} \rightarrow 0$ in the limit $\sigma_2 \rightarrow 0$, except the region near $x = 1$. It means that all these formulas obtained in FMSA approximation are not valid in the limit $\sigma_2 \rightarrow 0$. This is confirmed by the appearance of the divergencies in the expansions of σ_{eff} in small concentrations in the limit $z \rightarrow 1$. This fact is a consequence of the made approximation. For example, in case of the model of the layered squares this is due to the "closing" or "locking" effect of the layered structure in the adopted approximation in the limit $\sigma_2 \rightarrow 0$. In order to obtain a finite conductivity in this model above threshold concentration x_c one needs to take into account the correlations between adjacent squares. It is easy to show that near the threshold an effective conductivity is determined by random conducting clusters formed out of the crossing random layers from neighboring elementary squares. As is well known, the mean size of these clusters diverges near the percolation threshold [14, 15] and for this reason the FMSA approximation cannot be applicable for the description of σ_{eff} in the limit $z \rightarrow 1$ ($\epsilon > 0$) and of the percolation problem. It follows from figs. 3 that the EM approximation overestimates σ_{eff} , and both other formulas underestimate it in the region $z \rightarrow 1, \epsilon > 0$. We hope to investigate this limit in detail in the subsequent papers.

Thus we have discussed the possible functional forms of the effective conductivity of the two-phase system at arbitrary values of concentrations. A new functional equation, generalizing the duality relation, was deduced in the FMSA approximation and its solution was found. We have constructed also a hierarchical model of the random inhomogeneous medium and have found its effective conductivity in the same approximation at arbitrary phase concentrations. All formulas for the effective conductivity have the different functional forms. They

- (1) satisfy all symmetries including the dual symmetry and all necessary inequalities,
- (2) reproduce the general formulas for σ_{eff} in the weakly inhomogeneous case.

All these results confirm a conjecture that, in general, σ_{eff} of the two-phase randomly inhomogeneous system may be a nonuniversal function and can depend on some details of the structure of the randomly inhomogeneous regions. Analogous conclusions were done in paper [16] during a discussion of the possibility to find the generalization of the formula (3) for case $N \geq 3$.

The obtained formulas can be used for the approximation of the effective conductivity of some real randomly inhomogeneous systems like a corresponding formula of the EM approximation. The introduced composite method of the construction of the model random medium can be generalized on the other ways of determination of the effective intermediate conducting boxes. It can be done for different types of boxes as well as for different numbers of the possible types of the boxes. It is clear that then one will have to use another formulas instead of (3).

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